## USING THE LIGHT SCATTERING FUNCTION FOR STUDYING SUSPENDED MATTER IN THE SEA

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# USING THE LIGHT SCATTERING FUNCTION FOR STUDYING SUSPENDED MATTER IN THE SEA

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The angular distribution of the light scattered by sea water and its intensity are determined by the quantity, the material, the dimensions and the shape of the particles suspended in the sea water. By measuring the light scattering functions (phase functions), we can "transform" optical information, i.e., calculate the size distribution of the particles suspended in sea water from the optical data obtained. From a mathematical point of view the solution of this inverse problem reduces to one of solving the integral equation

$$\beta(\gamma) = \int_{r_1}^{r_2} \beta(\gamma, r) f(r) dr; \quad r_1 \leqslant r \leqslant r_2.$$
(1)

In this equation f(r) is the unknown particle size distribution;  $\beta(\gamma)$  is the measured scattering index as a function of the scattering angle  $\gamma$ ;  $\beta(\gamma, r)$  is the scattering factor of a nonodisperse suspension of particles of radius r (the kernel of the integral equation). Generally speaking, the stated problem is poorly conditioned in terms of the stability of the solution; however, for some limitation of the class of allowable solutions the inverse mapping  $\beta(\gamma) \rightarrow f(r)$  turns out to be stable [1]. For systems, consisting of arbitrary particles, when the kernel of the integral equation  $\beta(\gamma, r)$  is represented in the form of a table, there are various numerical methods of solving Eq. (1) [1-3]. For certain special cases, when a monodisperse scattering function can be expressed by a simple analytic formula, analytic methods of transforming the integral equation (1) have been developed in [1-7]. The use of these methods for studying suspended sea matter is very promising; however, certain difficulties, associated with the specific suspended sea matter, arise in the conversion of the optical information.

<sup>\*</sup>Numbers in the margin indicate pagination in the foreign text.

The particles suspended in sea water are extremely diverse in their composition and origin: They include terrigenous particles, brought into the sea by rivers or winds, phytoplankton cells, bacteria, particles of volcanic or even cosmic origin, and detritus—the remains of decomposed phytoplankton cells and the skeletons of zooplankton organisms. Zooplankton itself (to say nothing of fishes) in those concentrations in which it is found in sea water, exerts no marked effect on light scattering.

Suspended sea matter is a collection of particles with different refractive indices. Fortunately, the particles suspended in the sea are bounded to some extent in size and limited in their refractive indices. The formation of the size spectrum of the particles suspended in the sea is determined by settling and dissolution. The rates of both processes are strongly dependent on the sizes of the particles: settling slows down as the particles become smaller; the relative rate of dissolution, on the other hand, increases with a decrease in size. Organic and mineral particles have a different density. Whereas it is close to 1 g/cm3 for organic particles, it is two-three times higher for mineral particles. The heavy mineral particles settle faster than the organic. Particles of fine sand, 125 µm in diameter, settle at a rate of 1040 m/day; particles with a diameter of 16 µm settle at 19 m/day. For particles with sizes of 1-2 µm the settling rates are much less: 0.3 m/day for 2- $\mu$ m diameter, 0.07 m/day for 1- $\mu$ m diameter [8]. It is obvious that such particles can propagate over huge distances in the ocean, whereas the larger ones se tle out near the slore. Geologists point out that in open portions of the ocean most of the mass of terrigenous suspended matter enters into the finest fraction; findings of terrigenous sus ended matter greater than 1-µm in size are rare [9]. At the same time organic particles, on the other hand, are greater than 1  $\mu m$  in size: diatomic algae - 10-50, foraminifera - greater than 50, coccoliths - 5-15, organic detritus — 1-20  $\mu$ m [9].

What has been said above allows one to assume that particles having radii less than 1-2  $\mu$ m are primarily mineral; larger size particles are organic. For the mineral particles let us assume an average refractive index value of 1.15 [10]; for the organic it is less than 1.05\* [11].

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<sup>\*</sup>Studies of the absorption of filtered and unfiltered samples of sea water, conducted during the 5-th voyage of the scientific research vessel "Dmitriy Mendeleyev", showed that light absorption by the suspended particles is caused primarily by phytoplankton pigments. Outside the regions of absorption by the pigments the imaginary part of the refractive index can be ignored and the particles assumed to be transparent.

The shape of the suspended particles is immensely important. The fact of the matter is that for nonspherical particles there have been practically no calculations of the light scattering functions and factors. We are forced to assume the particles suspended in the sea are spherical since this is the only case of the stated problem that can be solved. We will follow the lead of geologists, who, by estimating particle sizes under a microscope, usually assign some one "average" size to the particles, assuming them to be spherical. In [12] it is pointed out that although diffraction by an isolated nonspherical particle depends heavily on its shape, the forward scattering for a system of nonspherical randomly oriented particles is not markedly different from the scattering by a system of spherical particles having projection areas equal to the average projection areas of the nonspherical particles. G. Beardsley [13], studying the light scattering matrix of sea water, concluded that the suspended sea matter behaves, in optical respects, like a collection of spherical particles. The question of the relationship between the geometrical and the defined effective dimensions remains unanswered.

Let us divide the entire size spectrum of the suspended sea particles into two fractions: 1) coarse — particle radius greater than  $r_2 = 1-2 \mu m$ ; 2) fine — particle radius less than 1-2  $\mu m$ . Let us represent the scattering factor  $\beta(\gamma)$  of sea water in the form

$$\beta(\gamma) = \beta_{w}(\gamma) + \beta_{f}(\gamma) + \beta_{c}(\gamma). \tag{2}$$

where  $\beta_W(\gamma)$  is the scattering factor of pure water;  $\beta_f(\gamma)$  is the scattering factor of the fine suspended matter;  $\beta_C(\gamma)$  is the scattering factor of the coarse particles. The values of  $\beta_W(\gamma)$  are known in [14]; therefore we can immediately subtract these values from the measured sea water function and then we only have to deal with the scattering function of the suspended matter.

The other two terms play a different role in different ranges of angles. An analysis of the correlation matrices of the scattering functions of sea water [15] shows that scattering at small angles is weakly correlated with the scattering at angles greater than 15°. This provides a basis for assuming that scattering at small and large angles is determined by two different, independent, fractions of sea water.

In sea water, as a rule, the fine suspended matter is heavily predominant; it usually amounts to more than 50% of the total quantity. According to M.V. Klenova's data, for the northern portion of the Atlantic ocean the particles less than 1  $\mu$ m in size

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amount to 60-86% of all the particles, 1-5  $\mu$ m — 13-39%, larger than 5  $\mu$ m — 0.3-2.1% [16]. These data agree qualitatively with the data of M.S. Barash [17] (also for the Atlantic ocean) and Yu.A. Bogdanov [18] (for the Pacific ocean). The coarse particles produce a scattering envelope that is severely elongated in the forward direction [19, 20]. Because of the fact that there are few of them suspended in sea water, their contribution at angles larger than 15° can be ignored compared with the scattering by the fine suspended matter. Let us first consider the interval of angles from 15 to 145° (where  $\beta_{\rm C}(\gamma)\approx 0$ ) and use the scattering function in this interval of angles for studying the quantitative and qualitative composition of the suspended matter in the sea. Then, after calculating the contribution of this fraction at small scattering angles, we determine the distribution of the coarse suspended particles in terms of size and number from the values of the scattering function at angles less than 10°, using the small-angle method of [4, 5].

#### MEASUREMENT METHOD

Two instruments are used for measuring the scattering envelopes of sea water; a visual spectro-hydronephelometer transmission meter (SHN) and an objective measuring device for small-angle scattering. The SHN instrument has already been described in the literature [21, 22]; we will give only some of its characteristics. The range of scattering angles measured is from 0.5 to 145°. The relative error for small scattering angles is 12%; for large angles it is 7%. The angular resolution of the instrument in the interval from 0.5 to 2.5° amounts to 0.5, from 2.5 to  $15^{\circ} - 2.5$ , from 15 to  $145^{\circ} - 4^{\circ}$ . As seen from the data presented, the SHN instrument does not provide measurements of high quality at small scattering angles; therefore a special scattering measuring device was used at these angles.

The concept of the measurement method was proposed in [4, 5], and a specific instrument for measurements in natural and artificial fogs, based on this principle, was constructed in [23] and studied in [24-28].

At the Institute of Oceanology this instrument was modified for measurements with sea water. Its optical system is shown in Fig. 1. Light from the light source 1-a K 12/50 incandescent lamp — is directed onto the small diaphragm 5 by means of the condenser 2. A stepwise optical attenuator 3 with neutral density filters makes it

possible to attenuate the flux from the source by factors of 10 and 100. The modulator 4, consisting of a G-33 motor and a disk with holes, modulates the light with a frequency of 1000 Hz. The interference light filter 6 makes it possible to operate in a narrow spectral interval ( $\lambda$  = 546 nm). The diaphragm 5 is located in the focal plane of the collimator 7. The divergence of the beam (in air) beyond the collimator amounts to 4'. The iris diaphragm 8 limits the beam diameter (d = 2 cm). The beam enters the cell 10 with the water to be tested through the input illuminator 9. The output illuminator serves as the objective of the detector 11. It is a plano-convex lens with the flat side facing the water. A photomultiplier with a point diaphragm 12 scans in its focal plane, measuring the irradiance distribution in this plane. An FEU-38 photomultiplier, specially selected from several samples, was used. The a-c electrical signal from the photomultiplier was fed to a V6-4 selective amplifier, and from it to an EPP-09 recorder (through a transformer). A seven-order-of-magnitude change in the light signal could be detected by electrical (by changing ranges on the V6-4) and optical (by means of the neutral density filters 3) methods. The signal, as a rule, exceeded the noise level by no more than an order of magnitude.

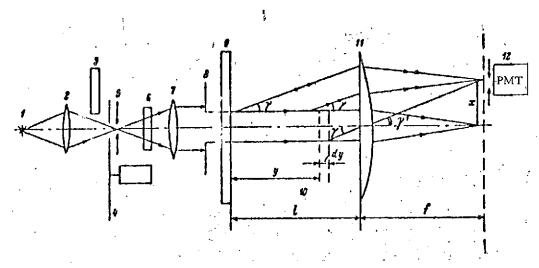


Fig. 1. Optical system of "small-angle" instrument. Explanation in text.

The received signal was recorded on the recorder chart; then the trace was analyzed for the purpose of calculating the absolute values of the scattering factor  $\beta(\gamma)$ . The relationship between the scattering factor  $\beta(\gamma)$  and the ratio of the light beams incident on the detector for an arbitrary and zero positions is expressed by the following formula:

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$$\beta(\tau) = \frac{\frac{4\pi f^0 m_0}{l S_{fs}} \left(\frac{\Phi_{\Upsilon}}{\Phi_0}\right)}{l S_{fs}}.$$
(3)

Here f is the focal length of the receiver lens 11; l is the length of the cell 10;  $S_{fs}$  is the area of the focal spot from the direct beam;  $m_0$  is the refractive index of sea water. Assuming that the coefficient of conversion of the light flux incident on the detector into recorder readings is the same for any scattering angle (including  $\gamma=0$  too), one can replace the light flux ratio  $\Phi_{\gamma}/\Phi_0$  by the ratio of recorder readings

$$\beta(\gamma) = \frac{4\pi f^2 m_0^2}{t S_{fs}} \left(\frac{n_1}{n_0}\right). \tag{3a}$$

The cell length l in our instrument was 0.24 m for operation in turbid waters and 0.51 m for clear waters. The focal length f of the receiver lens was equal to 393 mm; the area of the focal spot was  $S_{fs} = 0.61 \text{ mm}^2$ .

The scattering angle  $\gamma$  was determined from the recorder chart. The zero setting of the detector on the recorder chart is easily found from the signal maximum. Knowing the velocity of the carriage with the photomultiplier and the recorder chart speed, it is easy to relate the recorder chart displacement from the zero position to the scattering angle  $\gamma$ 

$$\gamma = \frac{0.85v_{\text{car}}}{v_{\text{ch}}f} \Delta L.$$
(4)

Here  $\theta_{\rm Car}$  is the velocity of the carriage with the photomultiplier;  $\theta_{\rm Ch}$  is the recorder chart velocity;  $\Delta L$  is the length of the chart from the zero position to the point of the reading; the coefficient 0.85 takes account of the refraction of the rays as they pass from water into air. Two PMT scanning velocities were used: 0.67 arc min/sec for covering the angular range from 0 to about 1° (the region where the scattering function changes most abruptly) and 1.92 arc min/sec for covering the rest of the angles. The moment of switching the scanning velocities was noted on the recorder chart. Upon traversing the entire range of angles, the carriage with PMT stopped automatically. The displacement of the carriage was monitored by means of a pointer instrument on the front panel of the detector unit.

The irradiance distribution in the focal plane of the receiver lens is determined not only by the light scattered by sea water, but also by all possible flashes, stray scattering, etc. In order to exclude this "background" from the measured distribution, the so-called "zero distribution" must be measured beforehand by filling the cell with "optically pure" water, i.e., with water that has been purified to the maximum possible extent of all absorbing inclusions (forward scattering can be ignored in this water). To accomplish this the sea water was filtered through a membrane filter with 0.15- $\mu m$  pore size and the ratio  $n_{\gamma}/n_0$  was measured for the filtered sample. This procedure was repeated 10-15 times, and from the distributions obtained those were selected which had low values of the ratio  $n_{\gamma}/n_0$  (3-5 distributions). For the curves selected an average distribution was computed, which was termed the "zero" distribution. The maximum deviations of the selected distributions from the average did not exceed 5%. Figure 2 shows an example of the zero distribution thereby obtained, making use of the 5-th voyage of the scientific research vessel "Dmitriy Mendeleyev". It is seen from the figure that even for an angle of 20' the magnitude of the background amounts to 10<sup>-4</sup> of the direct signal. The zero distribution was then subtracted from the measured distributions.

The random error of the determination of the value of  $\beta(\gamma)$  can be estimated to be no worse than 10% of the measured value. The possible systematic errors, associated with errors in the optical system used, have been analyzed by V.I. Golikov [24, 27]. We used the formulas derived by him, taking into consideration the parameters of our optical system. A calculation showed that neither spherical aberration nor the actual imprecise setting of the detector diaphragm are important for the measurements. The effect of decentering of the optical axes of the illuminator and light detector is more complicated to evaluate. But since we were dealing with a long-focus objective and we were not interested in the absolute values of the measured intensities, the decentering can evidently also be ignored [24, 27].

To check on the absence of systematic errors the data obtained on the small-angle instrument were

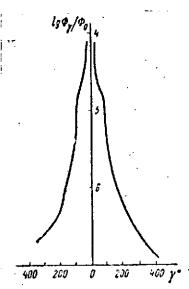


Fig. 2. Zero distribution for "small-angle" instrument (from measurements during 5-th voyage of the scientific research vessel "Dmitriy Mendeleyev").

joined with the data from the SHN instrument. The discrepancy between the data from both instruments in the angular interval from 2.5 to 7° did not exceed 10% as a whole, and in those cases when it reached greater values, it can nearly always be explained by a change in the samples due to the fact that they were not measured at the same time. At angles less than 2.5° the small-angle instrument deserves greater confidence since for the SHN the measurement field in this range of angles is generally nonuniformly illuminated, flashing is comparable to the quantity being measured and the accuracy of the  $\beta(\gamma)$  determination is not high. The good agreement of the absolute values of  $\beta(\gamma)$ , obtained by the two independent instruments, is yet another proof of the absence of  $\frac{1}{32}$  systematic errors in our measurements.

The size of the receiver diaphragm of the instrument was equal to the diameter of the focal spot, i.e., the detector aperture was also 4'. The angular resolution of the instrument was equal to 8' in air and smaller (7') in water. The minimum angle of the measurements was also equal to 7'; however, because of technical aspects of the instrument (the necessity of switching ranges on the V6-4 instrument) the actual minimum angle, for which reliable values could be obtained, was 20'.

## DETERMINATION OF THE COMPOSITION OF THE FINE SUSPENDED MATTER

As stated above, in the fine suspended matter we include particles whose radius is less than 1-2  $\mu$ m. Their refractive index is assumed equal to 1.1. The optical properties of these particles differ little from the optical properties of the surrounding medium; we will use the "soft" particle approximation for them [19, 20].

A method of inverting Eq. (1) — the so-called total indicatrix method — was developed and investigated in [6, 7] for these particles. In this method it is necessary to find the asymptotic form of the function  $g(x/2) \approx \beta(\gamma)/\psi(\gamma)$  to obtain the solution, where  $\psi(\gamma) \approx (1 + \cos^2\gamma)/(1 - \cos\gamma)^2$ ;  $x'/2 \approx \sin(\gamma/2)$ . The method imposes certain limitations on the particle size spectrum of the system being examined; the method cannot be used when a large number of fine (close to Rayleigh) particles are present. This case obviously occurs for exactly those suspended sea matters we have studied — the functions g(x/2) calculated for the measured scattering functions did not deviate from asymptotic behavior. We had to forget about using the total indicatrix method and had to make use of a trial-and-error method for determining the composition of the fine suspended matter.

It was shown in [29] that the scattering functions of the suspended sea matter of the waters of the northern Indian ocean are well approximated by the theoretical function calculated for a Young type of particle distribution for n = 5 in the "soft" particle approximation [30]:

$$f(r) = Ar^{-5}, \quad r_{\min} \leqslant r < \infty;$$

$$f(r) = 0, \quad 0 < r < r_{\min}.$$
(5)

Theoretical functions, agreeing well with the experimental, were successfully selected in [29] for the four types of scattering functions of this region that have been developed. The weight content and granulometric composition of the suspended matter, calculated theoretically from the measured scattering function, agree adequately well with those geological data which existed in the literature for the region investigated.

A study of the scattering functions for other regions of the oceans of the world showed that the measured functions are well approximated, as a rule, in their middle portions by the theoretical functions calculated for Young type distributions for n=4, 5 and 6. At small and large angles, however, the theoretical curves usually lie below the experimental. The too small values of the scattering at small angles for the theoretical indicatrices, compared with the experimental, must clearly be explained by the "under-estimation" of the Young type distributions of coarse particles in suspended sea matter, and at large angles — by the underestimation of the numbers of fine particles. Subsequently we used the Young type distribution only for particles with radii from  $r_1$  to  $r_2$ , while the small-angle method is employed for coarse suspended particles ( $r > r_2$ ). We assumed the finer particles ( $r < r_1$ ) to be Rayleigh in nature; for these optical data allow one to find only the product of the concentration by the average of the sixth power of the radius:  $\Delta = (Nr^6)_R$ . Considering the 15-145° range of angles, we can write

$$\beta_{\text{susp}}(\tau) = \beta(\tau) - \beta_1(\tau) = N_f c (1 + \cos^2 \tau) z (n, r_1, r_2, \tau) + c (1 + \cos^2 \tau) \Delta.$$
 (6)

for the scattering index of the suspended matter. Here  $N_f$  is the concentration of the fine  $(r_1\leqslant r\leqslant r_2)$  suspended matter;  $c=\frac{32\pi^5(m-1)^2\,(m+1)^2}{\lambda^4(m^2+2)^2}$  is a known constant, depending on the light wavelength and the refractive index of the particles;  $cz(n_1, r_1, \gamma)$ 

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 $(1+\cos^2\gamma)$  is the scattering index of the fine suspended matter of the only concentration conforming to a Young type distribution with the appropriate n,  $r_1$  and  $r_2$  parameters. The problem reduces to one of determining four parameters:  $N_f$ , n,  $r_1$  and  $\Delta$  (beforehand we choose  $r_2$  to be equal to 1 or 2  $\mu$ m; this parameter has slight influence on the shape of the function in the range of angles being considered). We use 10 scattering angles: 15, 30, 45, 60, 75, 90, 105, 120, 135, 145°. Let us divide Eq. (6) by  $c(1+\cos^2\gamma)$  and subtract from the result the value of c for  $\gamma=90^\circ$ . We obtain the expression

$$W(\gamma) - W(90^{\circ}) = N_{\rm f} \left[ z \left( n, \ r_{\rm f}, \ r_{\rm 2}, \ \gamma \right) - z \left( n, \ r_{\rm f}, \ r_{\rm 2}, \ 90^{\circ} \right) \right]. \tag{7}$$

which contains one less unknown than Eq. (6) (the ratio  $\beta_{\text{SUSP}}(\gamma)/[c(1+\cos^2\gamma)]$  is represented by W( $\gamma$ )).

After dividing Eq. (7) by W(15°) - W(90°), we obtain the expression

$$\frac{W(\gamma) - W(90^{\circ})}{W(15^{\circ}) - W(90^{\circ})} = \frac{z(n, r_1, r_2, \gamma) - z(n, r_1, r_2, 90^{\circ})}{z(n, r_1, r_2, 15^{\circ}) - z(n, r_1, r_2, 90^{\circ})},$$
(8)

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which contains only two unknown parameters: n and r1.

For Young type distributions with n=4, 5 and 6,  $r_1=0.01$ -0.20  $\mu m$ ;  $r_2=1$  and 2  $\mu m$ ; m=1.15 and  $\lambda=0.350$ , 0.383, 0.408, 0.421, 0.448 and 0.484  $\mu m$  we calculated 720 theoretical scattering functions on a "Minsk-22" computer using the formula

$$\beta(\gamma) = \frac{9\lambda^{2}}{128\pi} \cdot \frac{(m-1)^{2}(m+1)^{2}}{(m^{2}+2)^{2}} \cdot \frac{1+\cos^{2}\gamma}{\sin^{6}\frac{\gamma}{2}} \times \left((n-1)r_{1}^{n-1}\frac{1}{1-\left(\frac{r_{1}}{r_{2}}\right)^{n-1}}\cdot i\right),$$
(9)

where

$$I = \int_{r_1}^{r_2} \frac{(\sin q - q\cos q)^2}{r^n} dr; \quad q = \frac{4\pi r}{\lambda} \sin \frac{\tau}{2} [19].$$

For these theoretical functions the values of  $z(n, r_1, r_2, \gamma)$  and

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 $\frac{z(n, r_1, r_2, \gamma) - z(n, r_1, r_2, 90^\circ)}{z(n, r_1, r_2, 15^\circ) - z(n, r_1, r_2, 90^\circ)} \quad \text{were calculated, which were compared with the measured values of } [W(\gamma) - W(90^\circ)]/[W(15^\circ) - W(90^\circ)] \quad \text{in order to select the parameters n}$  and  $r_1$ . Then we determined  $N_f = \frac{W(15^\circ) - W(90^\circ)}{z(15^\circ) - z(90^\circ)} \quad \text{and } \Delta = W(\gamma) - N_f z(\gamma) \quad \text{as well as}$  the average value of this difference for angles  $60-145^\circ$ . The value of the maximum relative deviation  $[\Delta(\gamma) - \overline{\Delta}]/W(\gamma)$  was used as the approximation criterion. The minimum value of this maximum relative deviation corresponds to the best approximation.

Figure 3 shows the theoretical scattering functions compared with the observed data. As is evident from the figures, theoretical functions that are a good approximation of the experimental can be chosen for the most diverse regions of the oceans of the world (Atlantic and Pacific Oceans, Black Sea, closed basins — the lagoon of the Tarawa Atoll and Kieta Bay). In total we have analyzed in this manner several hundred scattering envelopes for the waters of the Indian Ocean (its northern part), Atlantic Ocean (Sargasso Sea), Pacific Ocean (various regions), Black Sea. For most samples the optimum optical model of the suspended sea matter was the Young type distribution with n = 5; the Young distribution with n = 4 was more appropriate for the waters of the Black Sea and the surface layer of the Southern Tradewind current in the Pacific Ocean (in its eastern part where it is polluted by the effluent of muddy waters of the Peru Current). For the subsurface waters of the Sargasso Sea the observed curves are well approximated by the theoretical functions calculated for a Young distribution with n = 6. In nearly all cases the approximation errors did not exceed the experimental errors.

This attests to the fact that the Young distribution with n = 4, 5 and 6 is a successful optical model for suspended sea matter with particles ranging in size from 0.01-0.20 to 1-2  $\mu$ m, i.e., for terrigenous suspended matter. Geological data have recently appeared, based on the use of a Coulter counter (which is capable of determining particles with diameters from 0.5 to several hundred  $\mu$ m [31]), indicating that in the tropical areas of the Atlantic Ocean the particle dispersion function is expressed by some combination of Young distributions.

To verify the proposed optical model we made use of the spectral dependence of the scattering factor in a given direction  $\beta(\gamma, \lambda)$ . Angles of 10, 15 and 30° were chosen, i.e., that range of angles where the scattering index  $\beta(\gamma)$  is nearly entirely determined by the fine suspended particles. A calculation was made for the six wavelengths 350, 383, 408, 421, 448 and 484 nm, corresponding to the effective wavelengths of the color

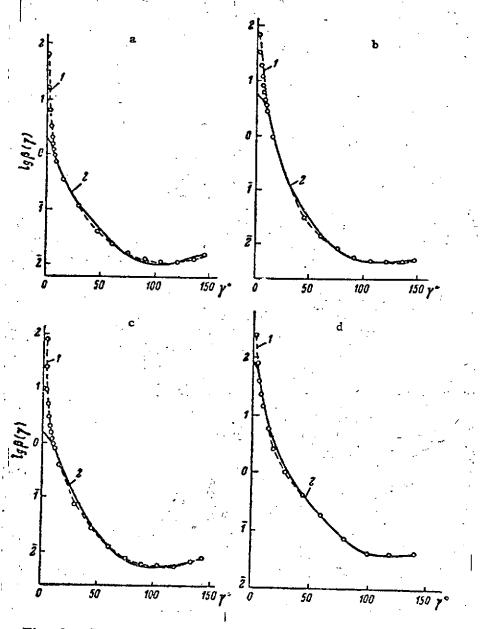


Fig. 3. Comparison of experimental (1) and theoretical (2) scattering functions. a) station 347, 500-m level, Sargasso Sea;  $\sigma=0.075~\mathrm{m}^{-1}$ ; matching parameters: n = 6, r<sub>1</sub> = 0.13  $\mu$ m, r<sub>2</sub> = 2  $\mu$ m; Nfine = 0.86  $\times$  10<sup>12</sup> m<sup>-3</sup>,  $\Delta$  = 2.1  $\times$  10<sup>-30</sup> m<sup>3</sup>; b) station 367, 5-m level, Pacific Ocean, Southern Tradewind Current;  $\sigma=0.16~\mathrm{m}^{-1}$ , matching parameters: n = 4, r<sub>1</sub> = 0.03  $\mu$ m, r<sub>2</sub> = 1  $\mu$ m, N<sub>f</sub> = 31  $\times$  10<sup>12</sup> m<sup>-3</sup>,  $\Delta$  = 0.27  $\times$  10<sup>-30</sup> m<sup>3</sup>; c) station 399, 10-m level, Pacific Ocean, southwest from island of Raratonga;  $\sigma=0.066~\mathrm{m}^{-1}$ ; matching parameters: n = 5, r<sub>1</sub> = 0.14  $\mu$ m, r<sub>2</sub> = 1  $\mu$ m, N<sub>f</sub> = 0.029  $\times$  10<sup>12</sup> m<sup>-3</sup>,  $\Delta$  = 0.89  $\times$  10<sup>-30</sup> m<sup>3</sup>;

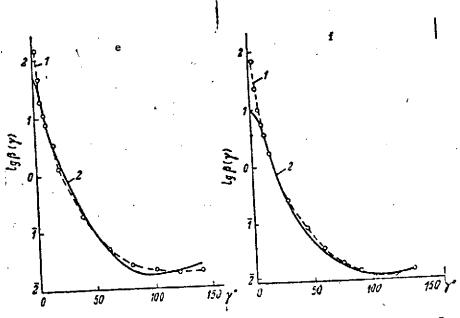


Fig. 3 (continued). d) Pacific Ocean, Kieta Bay (Solomon Islands), 5-m level;  $\sigma=0.54$  m<sup>-1</sup>, matching parameters: n=5,  $r_1=0.18$   $\mu m$ ,  $r_2$ , = 2  $\mu m$ ,  $N_f=1.0\times 10^{12}$  m<sup>-3</sup>,  $\Delta=3.6\times 10^{-30}$  m<sup>3</sup>; e) Pacific Ocean, lagoon of Tarawa Atoll (Gilbert Islands) 5-m level;  $\sigma=1.0$  m<sup>-1</sup>; matching parameters: n=5,  $r_1=0.10$   $\mu m$ ,  $r_2=2$   $\mu m$ ,  $N_f=17\times 10^{12}$  m<sup>-3</sup>,  $\Delta=4.5\times 10^{-30}$  m<sup>3</sup>; f) Black Sea, Karadag, 10-m level;  $\sigma=0.21$  m<sup>-1</sup>; matching parameters: n=4,  $r_1=0.15$   $\mu m$ ,  $r_2=1$   $\mu m$ ,  $r_3=1$   $\mu m$ ,  $r_4=1.6\times 10^{-30}$  m<sup>3</sup>.

Iters of the SHN instrument (taking into consideration the change in wavelength upon the transition from air to water). By way of an example Fig. 4 gives the results of a comparison of the calculated and measured functions  $\beta(10^{\circ}, \lambda)$ ,  $\beta(15^{\circ}, \lambda)$  and  $\beta(30^{\circ}, \lambda)$  for one sample of Black Sea water, taken in the vicinity of Karadag in the summer of 1969. For the  $10^{\circ}$  angle the theoretical and measured curves coincide within the measurement accuracy limits; for the  $15^{\circ}$  and  $30^{\circ}$  angles a discrepancy, in excess of the measurement error, is observed only at one point. The good agreement of the curves is a confirmation of the validity of our optical model.

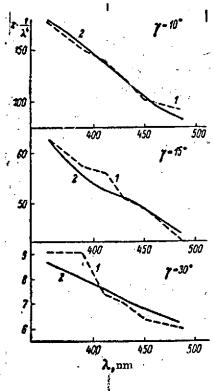


Fig. 4. Comparison of the experimental (1) and theoretical (2) wavelength dependence of the scattering factor in a given direction  $\beta(\gamma)$ , Black Sea, Karadag.

# DETERMINATION OF THE SIZE SPECTRUM OF THE COARSE SUSPENDED PARTICLES

We determined the size spectrum of the coarse suspended particles by the small-angle method [4, 5, 32]. Three modifications of the small-angle method have been developed in [32]: with differentiation of the measured function, from the current and from the integrated values of the scattering envelope. In order to compare the differentiation method and the current-value method a numerical experiment was performed. A model particle size spectrum was chosen (close to that which is usually found in suspended sea matter), for which the direct problem was solved, and then the resulting small-angle scattering function was inverted by two methods. The calculations were performed on a "Minsk-22" computer. A comparison of the computed spectra showed that the spectrum obtained by the differentiation method matches the model



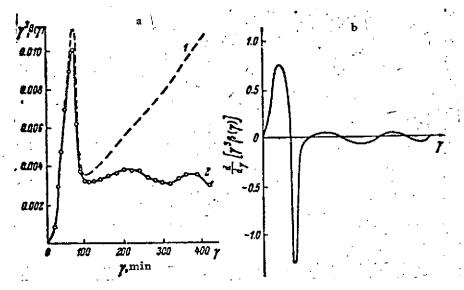


Fig. 5. The small-angle method. a) structure-sensitive function  $\gamma^3\beta(\gamma)$  for the scattering function of sea water (1) and coarse suspended matter (2); b) derivative  $d[\gamma^3\beta(\gamma)]/d\gamma$ .

spectrum better. Therefore this is the preferred method. The measured small-angle functions were inverted both manually (for the southwestern Pacific Ocean and the Black Sea [33]) and on the Minsk-22 computer (for the 5-th voyage of the scientific research vessel "Dmitriy Mendeleyev"). A specially conducted comparison of the manual and computer analysis showed good agreement of the results.

When the measured small-angle functions of sea water are directly inverted, the str cture-sensitive function  $\gamma^3\beta(\gamma)$  climbs upward steeply after the first minimum (Fig. 5, a). This is obviously due to the presence in the sea water of a large number of fine particles, falling outside the limits of the 2-30  $\mu$ m size range, which can be determined by the small-angle method [34, 35]. In this case we are forced to limit our consideration to only one period of change in the derivative (Fig. 5, b), i.e., to angles up to 2°. This is clearly insufficient for determining the particle size spectrum with good accuracy.

The simplest method is to get rid of the fine particles — to measure the scattering function not only for the sample as taken, but also for the sample filtered through a filter with 1-2- $\mu$ m pore radius. The difference in the scattering between the unfiltered and filtered samples must be due to the coarse suspended matter. Unfortunately, the contamination of the samples during filtering provides serious interference for the practical utilization of this method (in some cases the scattering of the filtrates was found to

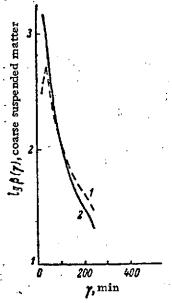


Fig. 6. Comparison of small-angle scattering functions of coarse suspended matter obtained as the difference between the scattering by unfiltered sample and by filtrate (1) and as the difference between the measured scattering and that calculated for the fine suspended matter (2).

be greater than that of the unfiltered samples). This contamination may be due to small dust particles entering the filtrate. Therefore we were forced to find the scattering function of the coarse suspended matter theoretically, by subtracting from the measured scattering function of sea water the scattering function of the fine suspended matter, calculated by the method discussed above. For a comparison Fig. 6 shows two small-angle functions of the coarse suspended matter for one and the same sample of Black Sea water. One of them was calculated theoretically while the second was obtained as the difference between unfiltered and filtered samples. An anomalous bend is observed on the second curve at small angles; the first curve is smooth. This difference can evidently be explained by the contamination of the filtrate by foreign (quite coarse) particles. The outstanding agreement of the curves in terms of absolute magnitude clearly indicates that there are no large errors in the method we have developed.

Such a method makes it possible to eliminate the rise in the structure-sensitive function beyond the

first minimum (Fig. 5, a) and to use three periods of variation in the derivative (Fig. 5, b) for the calculation, i.e., the interval of angles up to 7°. Figure 7 shows one of the size spectra for coarse suspended particles computed in this fashion. According to the estimates of V.I. Golikov, with 10% accuracy in the light measurements the maximum relative error (of the small-angle method) amounts to about 5-10% near the maximum of the distribution curve and increases to 25-40% at the base of this curve [36, 37]. The negative values obtained for f(r) at the right edge of the spectrum must be explained by incomplete optical information at very small scattering angles [34, 35].

To determine the absolute numbers of coarse suspended particles we normalized the size spectrum f(r) obtained by computing the total scattering factor of the coarse suspended matter as the difference between the total scattering function of sea water σ and the sum of the scattering factors of the fine suspended matter and pure water:

$$\sigma_c = \sigma - (\sigma_f + \sigma_w). \tag{10}$$

Here  $\sigma_{\rm W}$ ,  $\sigma_{\rm f}$ ,  $\sigma_{\rm c}$  are the total scattering factors of pure water, fine and coarse suspended matter, respectively. For the coarse particles the total scattering factor is equal to  $2\pi r^2$ , where r is the particle radius; one-half of this quantity is caused by geometrical optics and one-half by diffraction. For polydisperse suspended matter

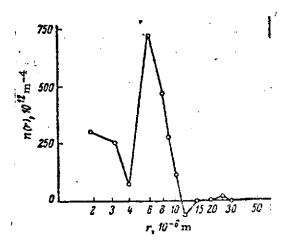


Fig. 7. Example of particle size spectrum calculated by small-angle method, southwestern portion of Pacific Ocean.

$$\sigma_{\rm c} = 2\pi N_{\rm c} \int_{\mathbf{r}}^{r_{\rm max}} f_{\rm c}(r) r^2 dr, \qquad (11)$$

from which we find the concentration of coarse particles

$$N_{c} = \frac{\int_{1}^{r} \sigma_{c}}{2\pi \int_{0}^{r} f_{c}(r) r^{2} dr}.$$
(12)

The coarse suspended particle distributions obtained in this manner agreed well with the fine suspended matter distributions both with respect to the relative behavior and also with respect to absolute values.

### COMPARISON WITH GEOLOGICAL DATA

Geologists gather suspended sea matter for study by two methods: with sample bottles followed by filtering of the sample through a membrane filter (usually filter No. 3 with 0.7- $\mu$ m pore size) and with separation by means of pumps while the vessel is sailing.

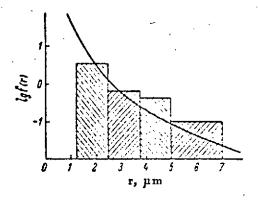


Fig. 8. Comparison of data on the size spectrum of suspended sea particles obtained by lightscattering method (curve) and by a direct counting of particles under a microscope (crosshatched areas), Black Sea, Karadag.

The suspended matter, collected on the filter, is subjected to microscopic analysis and, in addition, the filters are weighed to determine its weight concentration. Unfortunately, the standard method, employed by geologists and based on the use of conventional microscopes, does not permit an estimate of the number of very fine particles. The geological data are usually represented in the form of tables in which the number of particles is denoted by fractions: greater than 100, from 50 to 100, from 10 to 50, from 5 to 10, from 1 to 5, less than 1  $\mu$ m. The lower limit of the particle sizes determined depends on the

resolving power of the microscope, the individual characteristics of the eye of the observer and, as a rule, are not precisely known. The light-scattering method we have proposed has two important advantages over geological methods: 1) it makes it possible to estimate the number of very fine particles — up to 0.01-0.20  $\mu$ m (for the finer Rayleigh particles, as has already been stated above, the optical data determine only the product of the concentration and the average sixth-power of the particle radius (Nr  $^{\circ}$ )<sub>R</sub>); 2) it yields a continuous particle size distribution function in contrast to the discrete functions obtained by geologists. The accuracy of the standard geological method of microscopic analysis is not high and hardly exceeds the accuracy of particle size spectrum determination by the small-angle method.

It is natural that there is considerable interest in comparing data on the composition of suspended sea matter, obtained by the optical and geological methods. We have conducted such investigations on the Black Sea at the time of the Karadag expedition in the summer of 1969 and during the 5-th voyage of the scientific research vessel "Dmitriy Mendeleyev" in January-May 1971. In both cases one and the same water sample was selected for optical and for geological measurements. In the first case the particles under the microscope were counted by N.M. Nosenko, in the second case — by Yu. A. Bogdanov. Examples of the comparison of optical and geological data are shown in Fig. 8 for one of the Black Sea samples, and in the table for one of the samples taken during the 5-th voyage of the scientific research vessel "Dmitriy Mendeleyev" (station 399, 50-m level).

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Particle fraction (diameter), µm	Optical data, no. of particles	Geological data, thousand particles per liter
	2.8·10 <sup>5</sup> 1700 41 10 5.2 0.1	92.2 · 11.2 5.6 2,8 1.1

\*Smaller than 2.5 µm.

As is seen from the comparison that has been made, the optical data agree with the geological data in order of magnitude and likewise yield an overall identical size distribution behavior for suspended sea particles — a large predominance of fine particles.

As follows from the data presented, the number of fine particles is very large: the number of particles greater than 1  $\mu$ m in size amounts to less than 1% of the total number of particles. The concentration of suspended particles can be estimated at  $10^8-10^{10}$  particles per liter of sea water from the optical data, compared with  $10^4-10^6$  particles per liter from the geological data. The difference in the weight concentrations of the total amount of suspended matter and the suspended matter, collected by geologists, in the final analysis will not be too great, but nevertheless it can be assumed that separators collect only a small nortion of the total amount of suspended matter in sea water. Let us make an estimate using the optical data for the sample taken at station 399 from the 50-m level. Le us assume the density of the material of the fine suspended particles is 2.65 g/cm<sup>3</sup> [1t], and of the coarse suspended particles — 1.0 g/cm<sup>3</sup> [38]. The weight concentration C of the particles for any fraction with particle radii from  $\mathbf{r}_1$  to  $\mathbf{r}_1$  must be calculated from the formula

$$C = \frac{4}{3} \pi \rho N \int_{r_1}^{r_2} r^3 f(r) dr, \qquad (13)$$

where  $\rho$  is the density of the particle material, N is the total numerical concentration of suspended particles; f(r) is their size distribution.

The separator segregates the particles in terms of their hydraulic size; nevertheless, the lower limit on the radii of the sorted particles can be estimated as  $0.4 \,\mu\text{m}$ . Accordingly we calculate two quantities:  $C_1$  — the total weight concentration of the

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particles suspended in the sea water (in the radius interval from  ${
m r_1}$  to  ${
m r_{max}}$ ) and  ${
m C_2}$  the weight concentration of suspended matter sorted by the geologists (from  $r_{min}$  = 0.4  $\mu m$  to  $r_{max}$ ). Let us integrate Eq. (13) analytically from  $r_{min}$  to  $r_2$ , using Eq. (5); the integration from  $r_2$  to  $r_{max}$  is numerical, using the coarse particle size distribution density obtained by the small-angle method (station 399, 50-m level).

No. of particles, cm<sup>-4</sup> 13.104 4.0 2.0 1.4 0.26 0.13 0.080 0.064 0.048 0.016 < 0.001

The parameters of the Young type distribution for fine suspended matter were as follows: n = 5,  $r_1$  = 0.14,  $r_2$  = 2  $\mu m$ ;  $N_f$  = 0.28  $\times$  10<sup>6</sup> cm<sup>-3</sup>. A calculation yielded the following results:  $C_1 = 0.036$ ,  $C_2 = 0.014$  mg/liter. Thus, the separators gave less than onehalf the total quantity of suspended matter contained in sea water. This is the important result of our work, which, ultimately, still requires further checking.

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